



IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re Application of TORU HANADA, et al.
Serial No.: Application of 10/066,594
Filed: February 6, 2002
For: LIQUID CRYSTAL DISPLAY COMPONENT AND TRANSPARENT
CONDUCTIVE SUBSTRATE SUITABLE FOR THE SAME
Commissioner of Patents
Washington, D.C. 20231

DECLARATION

I, TORU HANADA, resides in 3-18-4 Tamadaira, Hino-shi, Tokyo-to, Japan, entered the Faculty of Materials Science and Engineering of Waseda University in April 1985 majoring in material science, took the master's course in electronics material science at the same University in March 1991, entered Teijin Limited in April 1991, was attached to the Polymer Material Research Laboratory of the Company, and engaged in research into improvement of the surface resistance of polycarbonate for 2 years, and has engaged transparent polymer films for a liquid crystal display.

As one of the inventors of the present inventors, I hereby resubmit my Declaration regarding US Patent No. 6,183,829. This Declaration consists of Part 1 and Part 2. Part 1 contains an added explanation regarding the previous Declaration submitted to the USPTO on May 14, 2003. Part 2 describes a new experiment demonstrating the superior properties (surface hardness and gas barrier properties) of the transparent polymer substrate of the present invention. The results in Part 1 and Part 2 show that our invention is in no way obvious from the prior art (Daecher, US 6,183,829 B1).

Part 1 (Additional explanation regarding previous Declaration submitted to USPTO on May 14, 2003)

In Run 2, a polycarbonate copolymer with a 50/50 (mole percent) composition of BCF and BisA and a viscosity average molecular weight of 37,000 (corresponding to the polymer described in Example 4 of the present patent specification) was used to produce a transparent polymer substrate by the solvent casting method. The brittleness was acceptable and the hue was satisfactory.

In Run 3, a polycarbonate copolymer having the same composition and the same viscosity average molecular weight as in Run 2 was used to attempt production of a transparent polymer substrate by an ordinary melt extrusion method, though different from the specified melt molding method described in the aforementioned prior art. The method described in the prior art, judging from Figs. 1 to 7 (particularly Fig. 7B), Claim 1 and the Examples, is a method of molding a film or sheet by allowing a molten resin heated at about 300°C to overflow and naturally flow downstream. The reason for using a melt extrusion method different from the prior art was that it is identical as a melt molding method and that we do not possess the specified apparatus described in the prior art.

We were not even able to obtain a film using the ordinary melt extrusion method. This was attributed to the high melt viscosity due to the large molecular weight of the polymer, which prevented extrusion of the polymer in Run 3.

In Run 1, therefore, we used a polymer having the same composition as in Runs 2 and 3 but a lower viscosity average molecular weight, for production of a transparent polymer substrate by the same melt extrusion method as in Run 3. However, as shown in Table 1, the resulting film had a yellow tint and was brittle, and thus was completely unusable as a transparent polymer substrate for displays.

We are therefore convinced that it is extremely difficult to mold, or impossible to obtain, a transparent polymer

substrate according to the present invention using a melt extrusion method similar to the method described in the prior art.

Part 2 (Confirming superior properties of transparent polymer substrate of the present invention)

1. Experiment

We assumed that of the polymers (particularly polycarbonates) described in the aforementioned prior art, "APEC DP9371" of Example 5 is closest to the polycarbonate copolymer forming the transparent polymer substrate of the present invention.

Using $^1\text{H-NMR}$ analysis, we determined that "APEC DP9371" is composed of a polycarbonate copolymer of IP: 1,1-bis(4-hydroxyphenyl)-3,3,5-trimethylcyclohexane and BisA: 2,2-bis(4-hydroxyphenyl)propane, with a compositional ratio (IP/BisA) of 60/40 (mole percent).

In the manner described below, we synthesized both the above-mentioned polycarbonate copolymer and our claimed polycarbonate copolymer of BCF: 9,9-bis(4-hydroxy-3-methylphenyl)fluorene and BisA: 2,2-bis(4-hydroxyphenyl)propane, in the same compositional ratio as "APEC DP9371", that is, (BCF/BisA) = 60/40 (molar ratio). We then fabricated a film with a thickness of 100 μm by the solvent casting method.

(1) Polymer synthesis and film fabrication

(Run 3): After placing 25,000 parts of ion-exchanged water and 4200 parts of a 48% aqueous sodium hydroxide solution in a reactor equipped with a thermometer, stirrer and reflux condenser and dissolving therein 4400 parts of 9,9-bis(4-hydroxy-3-methylphenyl)fluorene (hereinafter, "BCF"), 1100 parts of 2,2-bis(4-hydroxyphenyl)propane (hereinafter, "BisA") and 8 parts of hydrosulfite, there was further added 18,000 parts of methylene chloride, and then 2000 parts of phosgene was blown at 15-25°C while stirring, over a period of 60 minutes. After

blowing in the phosgene, a solution of 18 parts of p-tert-butylphenol dissolved in 330 parts of methylene chloride and 700 parts of a 48% aqueous sodium hydroxide solution were added for emulsification, after which 5.8 parts of triethylamine was added and the mixture was stirred for 1 hour at 28-33°C to completion of the reaction. The polycarbonate copolymer obtained by this procedure had a BCF/BisA constituent unit ratio (composition) of 60:40 as the molar ratio (polymer yield: 96%).

(Runs 1, 2, 4): The charging amounts of BCF and BisA were adjusted for the same method used above to synthesize polycarbonate copolymers with BCF/BisA compositional molar ratios of 30:70, 50:50 and 70:30.

These four polycarbonate copolymers were dissolved in methylene chloride to prepare solutions with polymer concentrations of 20 wt%. The solutions were then cast onto a polyester film to a thickness of 175 μm by the solvent casting method. Each coated film was heat dried to a residual methylene chloride content of 12 wt% in the film (at slightly different heating temperatures depending on the polymer composition), and then released from the polyester film. Each obtained film was further heat dried to fabricate a film having a residual methylene chloride content of 0.08 wt%. The film thickness was 150 μm .

(Run 5): The polymer "APEC DP9371" described in the aforementioned prior art was available from Bayer, Corp. A film with a thickness of 150 μm was then obtained by the solvent casting method the same manner as Run 3.

(2) Evaluation of gas barrier properties

The water vapor barrier property was evaluated by measuring the water vapor permeation rate in an atmosphere at 40°C, 90% RH using a PERMATRON W1A by Mocon.

The oxygen barrier property was evaluated by measuring the oxygen permeation rate in an atmosphere at 40°C, 90% RH using an OXTRAN 2/20ML by Mocon.

The results are shown in Table 2.

(3) Evaluation of pencil hardness

The pencil hardness of each of the obtained films was measured according to JIS K5400, using a Model HEIDON-14 by Shinto Scientific Co., Ltd. The pencil was affixed at an angle of 45° with respect to the film surface, a load of 1 kg was applied to the pencil in the direction normal to the film surface, and the film was shifted 1 cm in the horizontal direction at 30 mm/min, after which the presence of scratch marks was observed to evaluate the surface hardness. The results are shown in Table 2.

Table 2

Run No.	1	2	3	4	5
Sample Name	BCF30	BCF50	BCF60	BCF70	APEC9371
Substrate composition	BCF/BisA = 30/70	BCF/BisA = 50/50	BCF/BisA = 60/40	BCF/BisA = 70/30	IP/BisA = 60/40
Pencil hardness [JIS K5400]	H	H	H-2H	2H	B
Water Vapor Permeation Rate (g·100 $\mu\text{m}^2/\text{day}$)	46.1	37.7	35.6	31.9	91.5
Oxygen Permeation Rate (cc·100 $\mu\text{m}^2/\text{day}$)	930	630	470	320	4700

2. Results and Discussion

The results shown above demonstrated that the polycarbonate copolymer of the present invention (BCF/BisA) exhibited high pencil hardness (good>2H>H>B) and small permeation rates for water vapor and oxygen compared to the polymer described in the prior art. That is, the transparent polymer substrate of our present invention has highly superior pencil hardness and gas barrier properties.

In addition, all of the polycarbonate copolymer (BCF/BisA) compositions of 30/70 (Run 1), 50/50 (Run 2) and 70/30 (Run 4) exhibited excellent pencil hardness and gas barrier properties comparable to Run 3.

The polycarbonate copolymer of the present invention has

high surface hardness and gas barrier properties which are highly superior to those of the polymer "APEC DP9371" specifically described in the prior art, and it is indeed suitable as a film for displays. This confirms that the present invention can in no way be considered obvious from Daecher. We are therefore convinced that our invention possesses adequate patentability.

The undersigned declarant further declares that all statements made herein of his own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issuing thereon.

The 25th day of November, 2003



Toru HANADA